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Cross-correlation frequency-resolved optical gating by molecular vibration for ultrashort pulse

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The frequency-resolved optical gating (FROG) technique is one of the most popular and robust methods for ultrashort laser pulse characterization [1], which involves an experimental apparatus to create a spectrogram of a target pulse resulting from its convolution with a gate function and a retrieval algorithm.

Since a nonlinear frequency mixing crystal is required, the ultrashort pulse measurement is restricted by the achievable phase-matched spectral bandwidth, group velocity mismatch, frequency conversion efficiency, transparent spectral range of the crystal, and material dispersion induced broadening of the pulses. We propose cross-correlation frequency-resolved optical gating (XFROG) technology based on molecular vibration, which is not limited by the wavelengths, and also has high sensitivity for the measurement of the weak pulse.

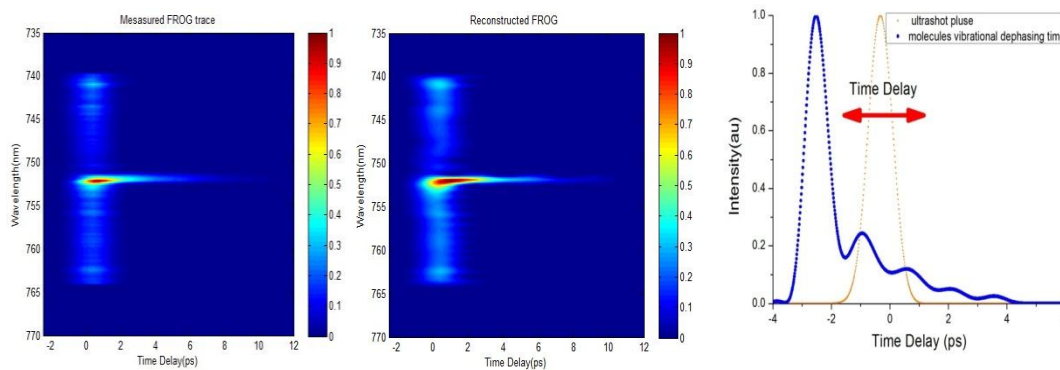


Fig. 1 (a) Measured XFROG trace and (b) reconstructed XFROG trace (c) ultrashort probe pulse and molecules vibrational dephasing time (blue line)

Coherent anti-Stokes Raman scattering (CARS) is a nonlinear four-wave mixing process, vibrational coherence is prepared when the pump and Stokes excitation fields overlap temporally and spatially on a Raman-active sample. The molecules vibration were impulsively stimulated by the laser pulses, which is then field-free revived with dephasing time determined by the molecular constants due to the quantum beatings of the molecular vibrations. The CARS signal results from the scattering of the probe pulse off the excited vibration coherence. We measured the intensity changes of CARS spectral signals for dimethyl sulfoxide versus delay time detected by our broadband time-resolved CARS with a 18cm PCF pumped by 783nm of wavelength, 150fs, 20kW laser pulse [2]. Fig. 1 (a) shows one of the T-CARS signals: the vertical axis corresponds to the CARS wavelength, which correlates to the wavenumber. The horizontal axis corresponds to the delay time between the probe and supercontinuum pulses. The process of vibrational coherence is mostly on ps or fs scale (0.5 ps ~10 ps) [3]. We used the molecular dephasing time as the transient gate function, which exhibited a process of exponential decay, and consequently led to a clear XFROG spectrogram. To retrieve the probe pulse from the measured XFROG trace, we implemented an iterative Fourier-transform algorithm with a generalized projection method. As we can see, the retrieved XFROG trace in the time domain, Fig. 1(c), shows the molecular vibrational dephasing time (blue curve) and the ultrashort pulse (yellow curve).

In summary, we have demonstrated that molecular-vibration-gated XFROG is a powerful technique for the measurement of ultrashort pulses and ultrafast molecular vibration. Since the molecular vibrational coherence time is from fs to ps, we can expect to measure different ultrashort pulse durations by choosing different Raman modes.

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